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A note on the dynamical structure of liquid LiNa alloy

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Abstract : The dynamical structure factors of liquid LiNa alloy are calculated in the visco-elastic approximation using static properties like effective pair potential and static structure factors The results are compared with inelastic neutron scattering (INS) and molecular dynamics (MD) simulation results The specific features are discussed It is clearly shown that the effect of the concentration variation on the dynamical structure is small though the corresponding effect on the static structure is quite large

Keywords : Dynamical structure, ordering potential, visco-elastic approximation.

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Based on the modified Young-Hoshino (MYH) potential for Li and simple Ashcroft model for Na, the successful calculation of Bhatia-Thornton (B-T) static structure factors and the thermodynamic properties of liquid LiNa system at different concentrations [1] and thereafter the dynamical properties like diffusion constants [2], we are tempted to see the applicability of this model for the calculation of dynamical structure factors of this system at different concentrations. This is important because, as pointed out by Sinn *et al* [3], the precise measurements of the dynamical structure factors provide a stringent test of the inter-atomic interactions. For instance, the results of dynamical structure factors for liquid Li obtained with neutral pseudo-atom method [4] compare well with their experimental data, whereas Ashcroft's empty core model appears to be inadequate. Evidently the choice of the model potential is important.

The liquid LiNa alloy is a typical phase separating (ps) system whose stoichiometric composition (c_{Li} = 0.61) is also the null alloy composition in neutron diffraction. The total static structure factor, $S(q)$ reduces to concentration-concentration

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structure factor, $S_{cc}(q)/c_{Li}c_{Na}$ and is dominated by a strong small angle scattering with subsequent weak oscillations [1]. The system possesses a typical ordering potential. It is therefore interesting to see if these features in the ordering potential and static structure factor have appreciable effects on the dynamical structure.

For binary liquid alloy in the effective medium approximation (EMA) [5] we use the concept of Wigner-Seitz sphere treating them as a pseudo one component system with mean potential $(\phi_m(r) \equiv \phi_{NN}(r))$, obtainable from component pair potentials and the pure component parameters are replaced by effective values. For dynamical structure factor, $S(q,\omega)$ we consider visco-elastic approximation [6]. The visco-elastic model is simplest one to use and has been applied to a number of systems with reasonable success. It gives a method for connecting dynamical properties to static properties like liquid structure factor and effective pair potential. The method is based on phenomenological analysis of time correlation function and principal advantage is that it provides unified treatment of a wide variety of transport coefficients. In the model stress correlation functions can be related to intermediate dynamic scattering function and expression for $S(q,\omega)$ can be conveniently obtained. Thus, $S(q,\omega)$ is given by

$$S(q,\omega) = \frac{1}{\pi} \frac{\tau(q) v_{th}^2 q^2 \left[\omega_L^2(q) - \omega_0^2(q)\right]}{\left\{\left[\omega\tau(q)(\omega^2 - \omega_L^2(q))\right]^2 + \left[\omega^2 - \omega_0^2(q)\right]^2\right\}} \tag{1}$$

with

$$\frac{1}{\tau(q)} = \frac{2}{\sqrt{\pi}} \sqrt{\left(\omega_L^2(q) - \omega_0^2(q)\right)}, \quad \omega_0(q) = v_{th} q / \sqrt{S(q)} \tag{2}$$

where v_{th} is the thermal velocity and $\omega_L(q)$ is the longitudinal frequency of the associated wave. $S(q)$, the total structure factor for LiNa alloy can be obtained in the low order perturbation theory using appropriate pseudopotentials [1]. For $\omega_L(q)$ we follow Bhatia-Singh calculation procedure [7] primarily intended for amorphous systems. The relations can be extended to liquids and mixtures with some modifications. For liquid, the parameters β and δ of the first and the second derivatives of the effective pair potential are related and the modified dispersion relations for liquid can be obtained [7].

We consider LiNa liquid alloy at T = 725 K and ionic density, ρ = 0.031083 Å^{-3}. In Figure 1 we show the calculated $S(q)$ and $S_{cc}(q)/c_{Li}c_{Na}$ at three different concentrations. The general features of the experimental (neutron) $S(q)$ curves [8] are represented reasonably well by our model potentials as shown in our previous paper (Ref. [1] of this paper) for c_{Li} = 0.61. The agreement with other curves is similar. For c_{Li} = 0.61, $S(q)$ based on MD simulation [9] has also similar features. In the model,

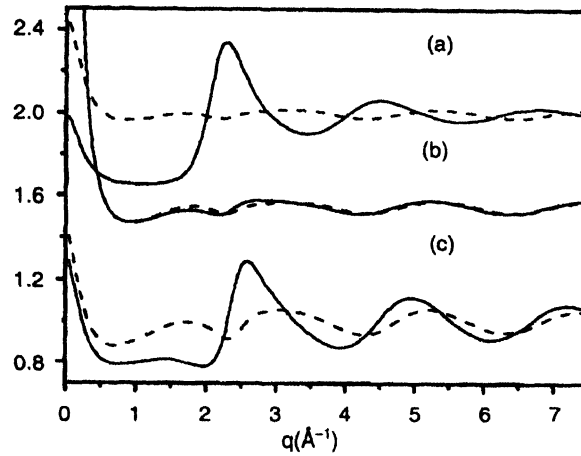

Figure 1. LiNa liquid alloy, structure factors *vs.* q : $S(q)$ ——, $S_{cc}(q)/c_{Li}c_{Na}$ – – – at (a) $c_{Li} = 0.3$ (origin shifted by 1.0) (b) $c_{Li} = 0.61$ (null alloy) (origin shifted by 0.5) (c) $c_{Li} = 0.8$.

at $c_{Li} = 0.61$, $S(q)$ and $S_{cc}(q)/c_{Li}c_{Na}$ are almost indistinguishable while for $c_{Li} = 0.3$ and $c_{Li} = 0.8$ they are very different in their features. The calculated $\omega_L(q)$ (not shown) appears in some respect similar to that of pure liquid Li.

The dynamical structure factors have been obtained for three q-values, $q = 0.8$, 1.2 and 1.5 Å^{-1} and the results are shown in Figure 2. The results when compared with MD simulation results [10], we find reasonably good agreement for $q = 0.8$ and 1.2 Å^{-1}. The experimental, INS data [11] are also shown and there is some agreement. The agreement is reasonable in the sense that the viscoelastic model is an appropriate model and no calculation even on liquid metals *e.g.* Li, Rb *etc.* could yield better agreement [12] with MD/experimental results. Further LiNa is a phase separating non-simple alloy where the application of the model is surely approximate. In this connection it is also to be noted that when the experimental, INS data is integrated over ω to give the static $S(q)$, the obtained results show some discrepancies with experimental $S(q)$ result [10] while MD data give a better agreement. Evidently, the experimental (INS), $S(q,\omega)$ data are somewhat lower. We have also done calculations for $c_{Li} = 0.3$ and 0.8, to see the concentration dependence of $S(q,\omega)$. At $q = 0.8$ and 1.2 Å^{-1}, the effect of concentration on $S(q,\omega)$ is small while at $q = 1.5$ Å^{-1} the effect is significant.

Though the total static structure factors are different considerably with concentration at these q-values $S(q,\omega)$ are not affected much. Again at lower concentration, $S(q,\omega)$ are mostly lower at $q = 0.8$ and 1.2 Å^{-1} while this trend is almost reversed at $q = 1.5$ Å^{-1}. At $q = 1.5$ Å^{-1}, however, our calculated $S(q,\omega)$ are relatively not in good agreement with MD as well as INS results. Anyway, it is clear that though the effect of concentration is enormous on static structure factor and ordering potential, it is less prominent on dynamical structure factor. The possible explanation

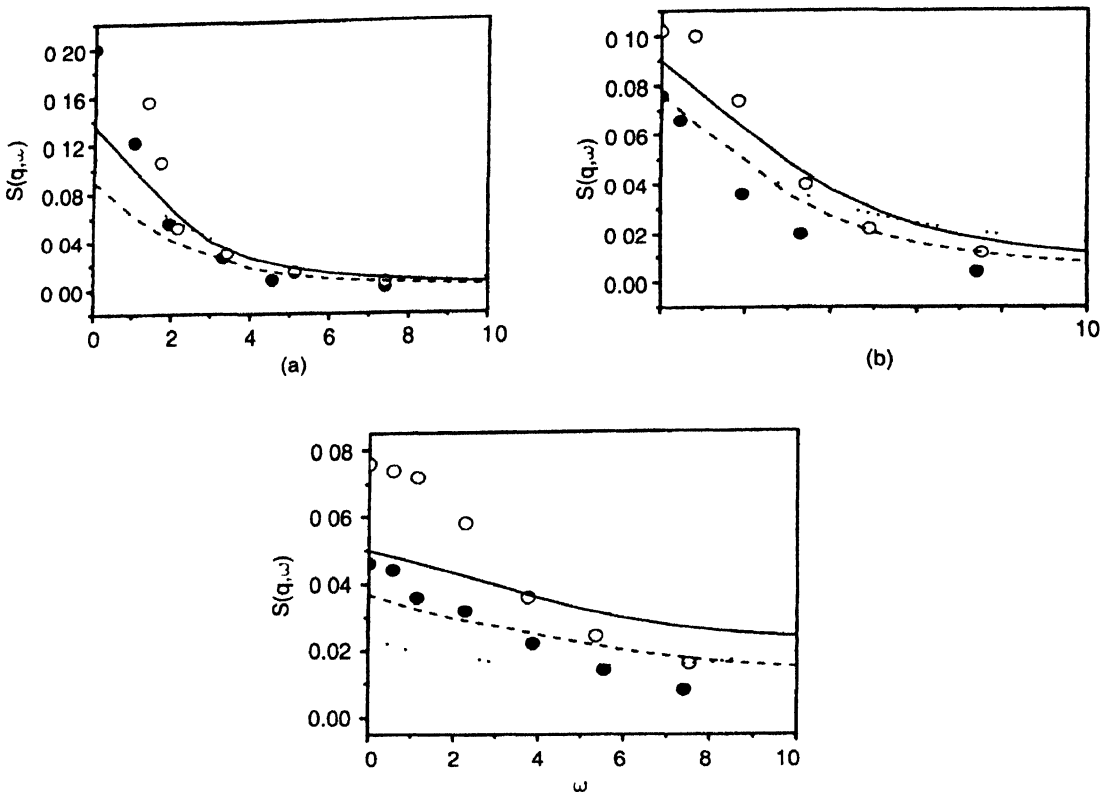

Figure 2. $S(q,\omega)$ *vs.* $\omega(\omega$ in $p\,s^{-1})$ for liquid LiNa alloy for (a) $q = 0.8$ Å^{-1}, (b) 1 2 Å^{-1} and (c) 1.5 Å^{-1}

Calculated — (at 0 61 Li concentration), – – – (at 0.3 Li concentration) and .... .. (at 0.8 Li concentration), MD simulation result ooo and experimental result ●●● both at 0.61 Li concentration

may be as follows. $S(q,\omega)$ in the explored (q,ω) region decreases monotonically as function of ω with no side peak. At null alloy (c_{Li} = 0.61), $S(q,\omega)$ is entirely given by $S_{CC}(q,\omega)$ which therefore follow the same pattern of $S(q,\omega)$. Though $S_{NN}(q,\omega)$ shows side peak [10] its contribution to $S(q,\omega)$ is very small at c_{Li} = 0.3 and 0.8. Again, like $S_{CC}(q)$ (Figure 1 except at $q \rightarrow 0$), $S_{CC}(q,\omega)$ is also expected to be not strongly concentration dependent. So it is expected that $S(q,\omega)$ is only weakly dependent on concentration. The concentration dependence is however prominent at larger Li-concentration and larger q-values. In case of static structure, however, the $S_{NN}(q)$ and $S_{CC}(q)$ have basic differences in shape. While $S_{NN}(q)$ has a pattern more like that of pure Li or Na with a large first peak, $S_{CC}(q)$ has a small double peak with low-q region changing enormously with concentration and this shoots up at C_{Li} = 0.61. This difference in $S_{NN}(q)$ and $S_{CC}(q)$ structure ($S_{NC}(q)$ is very small at all q) is responsible for large change in $S(q)$ with concentration. Further, the overall features of $S(q,\omega)$ curves are that unlike pure liquid Li, no side peaks revealing the existence of propagating sound modes are present.

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